Non-uniform convergence of two-photon decay rates for excited atomic states

Ulrich D. Jentschura

Max–Planck–Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

Abstract Two-photon decay rates in simple atoms such as hydrogenlike systems represent rather interesting fundamental problems in atomic physics. The sum of the energies of the two emitted photons has to fulfill an energy conservation condition, the decay takes place via intermediate virtual states, and the total decay rate is obtained after an integration over the energy of one of the emitted photons. Here, we investigate cases with a virtual state having an energy intermediate between the initial and the final state of the decay process, and we show that due to non-uniform convergence, only a careful treatment of the singularities infinitesimally displaced from the photon integration contour leads to consistent and convergent results.

PACS numbers 12.20.Ds, 31.30.Jv, 06.20.Jr, 31.15.-p

Two-photon decay in atomic systems continues to be of both theoretical and experimental interest today (see, e.g., Ref. [1]). Here, we shall investigate a mathematical subtlety of the problem, which ultimately reveals that correct results for two-photon decay widths of highly excited states depend on a careful analysis of the singularities close to the photon integration contour. In general, the decay width of a bound system may be understood naturally as the imaginary part of the self energy [2]. This because the fundamental time evolution of a Schrödinger eigenstate of energy E_i, which reads $\exp(-\mathrm{i} E_i t)$, must be modified to $\exp[-\mathrm{i}\,(E_i + \mathrm{Re}\,\Delta E_i)\,t - \frac{1}{2}\,\Gamma\,t]$ once a perturbation leads to both a real and an imaginary energy shift according to

$$E_i \to E_i + \Delta E_i\,, \qquad \Delta E_i = \mathrm{Re}\,\Delta E_i - \mathrm{i}\,\frac{\Gamma}{2}\,. \tag{1}$$

It is known [3] that the imaginary part of the two-photon self-energy shift $\mathrm{Im}\Delta E_i = -\frac{1}{2}\,\Gamma$ gives rise to the two-photon decay width. Important steps toward a full clarification of the two-photon processes involving excited states have been accomplished in Refs. [4–6]. Here, it is our intention to clarify the role of intermediate, virtual states, whose energy lies between the energy E_i of the initial state and the energy E_f of the final state of the decay process. The concept developed in [2] guides us in our investigation.

We start from the nonrelativistic two-loop self energy [3, 7] in natural units ($\hbar = c = \epsilon_0 = 1$), with m denoting the electron mass and α the fine-structure constant,

$$\Delta E_i = \lim_{\epsilon \to 0} \left(\frac{2\alpha}{3\pi m^2}\right)^2 \int_0^{\Lambda_1} d\omega_1\,\omega_1 \int_0^{\Lambda_2} d\omega_2\,\omega_2\, f_\epsilon(\omega_1, \omega_2)\,. \tag{2}$$

Here, Λ_1 and Λ_2 are ultraviolet cutoff parameters, and in $f_\epsilon(\omega_1, \omega_2)$ we carefully keep track of all infinitesimal imaginary parts,

$$\begin{aligned}
f_\epsilon(\omega_1,\omega_2) = {} & \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_1 + \mathrm{i}\epsilon}\, p^k\, \frac{1}{E - H - \omega_1 - \omega_2 + \mathrm{i}\epsilon}\, p^j\, \frac{1}{E - H - \omega_2 + \mathrm{i}\epsilon}\, p^k \right| \phi_i \right\rangle \\
& + \frac{1}{2} \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_1 + \mathrm{i}\epsilon}\, p^k\, \frac{1}{E - H - \omega_1 - \omega_2 + \mathrm{i}\epsilon}\, p^k\, \frac{1}{E - H - \omega_1 + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle \\
& + \frac{1}{2} \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_2 + \mathrm{i}\epsilon}\, p^k\, \frac{1}{E - H - \omega_1 - \omega_2 + \mathrm{i}\epsilon}\, p^k\, \frac{1}{E - H - \omega_2 + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle \\
& + \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_1 + \mathrm{i}\epsilon}\, p^j\, \left(\frac{1}{E - H}\right)'\, p^k\, \frac{1}{E - H - \omega_2 + \mathrm{i}\epsilon}\, p^k \right| \phi_i \right\rangle \\
& - \frac{1}{2} \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_1 + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle \left\langle \phi_i \left| p^k \left(\frac{1}{E - H - \omega_2 + \mathrm{i}\epsilon}\right)^2 p^k \right| \phi_i \right\rangle \\
& - \frac{1}{2} \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_2 + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle \left\langle \phi_i \left| p^k \left(\frac{1}{E - H - \omega_1 + \mathrm{i}\epsilon}\right)^2 p^k \right| \phi_i \right\rangle \\
& + m \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_1}\, \frac{1}{E - H - \omega_2 + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle - \frac{m}{\omega_1 + \omega_2} \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_2 + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle \\
& - \frac{m}{\omega_1 + \omega_2} \left\langle \phi_i \left| p^j \, \frac{1}{E - H - \omega_1 + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle .
\end{aligned} \tag{3}$$

Here, E_i is the Schrödinger energy of the reference state, which is qualified here as the initial state of the two-photon decay process, and H is the Schrödinger Hamiltonian. Sums over the Cartesian coordinates $j, k \in \{1, 2, 3\}$ are implied throughout this communication (summation convention). One may ask why the infinitesimal imaginary parts in the propagator denominators have such a sign that E_i effectively seems to acquire a positive

imaginary part. That is not the case: the reference state is assumed to be an asymptotic state in this formalism and does not have any imaginary part associated to it at all. The infinitesimal imaginary parts are due to the virtual states (included in H), and these acquire an infinitesimal negative imaginary part, as they should.

If the reference state is an excited state, then various singularities are encountered along both the ω_1 and ω_2 integrations. As shown in Ref. [3], the two-photon decay rate can be obtained from the imaginary part of the two-loop self energy, upon consideration of those imaginary parts which are generated when a virtual state $|\phi_v\rangle$ with energy E_v and the two emitted photons meet at a resonance condition: $E - E_v = \omega_1 + \omega_2$. At these points, expressions of the type $1/(E - H - \omega_1 - \omega_2)$ become singular.

In order to allow for a consistent treatment of the two-photon decay rate of excited states, it is necessary to treat the energy shift (2) as a whole, to carefully keep track of all $\mathrm{i}\epsilon$ terms, and to defer the distinction of imaginary and real parts to a later point. Carrying out the integration over one of the photon energies using the Dirac prescription

$$\frac{1}{a - \omega + \mathrm{i}\epsilon} = -\mathrm{i}\pi\,\delta(\omega - a) + (\mathrm{P})\frac{1}{a-\omega}\,, \tag{4}$$

where (P) denotes the principal value, we find that the two-photon decay rate corresponds to the expression

$$\frac{\Gamma}{A} = \lim_{\epsilon\to 0} \mathrm{Re} \int\limits_0^{E_i - E_f} \mathrm{d}\omega\, \omega\,(E_i - E_f - \omega) \left\{ \left\langle \phi_f \left| p^j \, \frac{1}{E_i - H - \omega + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle + \left\langle \phi_f \left| p^j \, \frac{1}{E_f - H + \omega + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle \right\}^2 . \tag{5}$$

where

$$A = \frac{4}{27}\,\frac{\alpha^2}{\pi}\,, \tag{6}$$

In general, the expression (5) has both a real and an imaginary part (where it not for the enforced selection of the real part implied by the "Re" in the cited equation). In that context, it is useful to observe that Γ already manifests itself as the imaginary part of the energy shift (2). The real part of Γ, in turn, gives the decay rate, and by consequence the "imaginary part of Γ" corresponds to a real part of the original energy shift (2), which is of the "squared decay-rate" type discussed in [8, 9]. The structure of the energy shift associated with a resonance is quite intriguing in higher orders.

In a basis-set representation, Eq. (5) reads

$$\frac{\Gamma}{A} = \lim_{\epsilon\to 0} \mathrm{Re} \int\limits_0^{E_i - E_f} \mathrm{d}\omega\, \omega\,(E_i - E_f - \omega) \sum_v \left\{ \frac{\langle \phi_f\, | p^j |\, \phi_v \rangle \, \langle \phi_v\, | p^j |\, \phi_i \rangle}{E_i - E_v - \omega + \mathrm{i}\epsilon} + \frac{\langle \phi_f\, | p^j |\, \phi_v \rangle \, \langle \phi_v\, | p^j |\, \phi_i \rangle}{E_f - E_v + \omega + \mathrm{i}\epsilon} \right\}^2 , \tag{7}$$

where the sum over v contains all virtual states, i.e. over the entire bound and continuous spectrum.

The expression (5) now gives us a clear prescription how to handle the potentially problematic case of a virtual state $|\phi_v\rangle$ having an intermediate energy E_v with $E_f > E_v > E_i$. An example is a virtual $|\phi_v\rangle = |2\mathrm{P}\rangle$ state for a reference state $|\phi_i\rangle = |3\mathrm{S}\rangle$ and a final state $|\phi_f\rangle = |1\mathrm{S}\rangle$. The treatment can be illustrated in a very clear manner by investigating the general structure of the terms generated by the virtual states with intermediate energies $E_f > E_v > E_i$. We treat the square of the first term in curly brackets in Eq. (7) as an example. Indeed, after an appropriate scaling of the photon energy integration variable, the expression takes the following form ($0 < a < 1$),

$$\lim_{\epsilon\to 0} \mathrm{Re} \int_0^1 \mathrm{d}\omega \left(\frac{1}{a - \omega + \mathrm{i}\epsilon} \right)^2 = \lim_{\epsilon\to 0} \int_0^1 \mathrm{d}\omega \, \frac{(a-\omega)^2 - \epsilon^2}{[(a-\omega)^2 + \epsilon^2]^2} = \lim_{\epsilon\to 0} \left(\frac{a-1}{(a-1)^2 + \epsilon^2} - \frac{a}{a^2 + \epsilon^2} \right) = \frac{1}{a(a-1)}\,. \tag{8}$$

This result holds strictly for $0 < a < 1$, but the limit is not approached uniformly, i.e. it would be forbidden to exchange the sequence of the limit $\epsilon \to 0$ with the integration over ω. Similar phenomena are observed in the context of the renormalization of quantum electrodynamic processes, which necessitate the preservation of all relativistically covariant regulators up to the very end of the calculation, also due to non-uniform convergence. Having Eqs. (5) and (7), it is easy to perform actual numerical evaluations of the two-photon decay rates for hydrogenlike systems. Numerical results are given in Table 1.

By contrast, let us suppose we had replaced the right-hand side of (5) by the expression

$$\int\limits_0^{E_i - E_f} \mathrm{d}\omega\, \omega\,(E_i - E_f - \omega) \left| \left\langle \phi_f \left| p^j \, \frac{1}{E_i - H - \omega + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle + \left\langle \phi_f \left| p^j \, \frac{1}{E_f - H + \omega + \mathrm{i}\epsilon}\, p^j \right| \phi_i \right\rangle \right|^2 , \tag{9}$$

Table 1: Two-photon decay rates obtained for hydrogenlike systems by evaluation of Eq. (5). Various excited nS states are considered, both as initial (excited) and as final states of the two-photon process. The results given here are for nuclear charge $Z = 1$ and scale with Z^6 for hydrogenlike systems. Units are inverse seconds. To obtain the decay rate in Hertz, one divides by a factor of 2π.

	$\vert\phi_i\rangle = \vert 2\mathrm{S}\rangle$	$\vert\phi_i\rangle = \vert 3\mathrm{S}\rangle$	$\vert\phi_i\rangle = \vert 4\mathrm{S}\rangle$	$\vert\phi_i\rangle = \vert 5\mathrm{S}\rangle$
$\vert\phi_f\rangle = \vert 1\mathrm{S}\rangle$	8.229 352	2.082 853	0.698 897	0.287 110
$\vert\phi_f\rangle = \vert 2\mathrm{S}\rangle$	–	0.064 530	0.016 840	0.001 809
$\vert\phi_f\rangle = \vert 3\mathrm{S}\rangle$	–	–	0.002 925	0.000 704
$\vert\phi_f\rangle = \vert 4\mathrm{S}\rangle$	–	–	–	0.000 297

which has no physical meaning. We have enforced a real valued integrand by the introduction of the complex modulus. In that case, we would have obtained a divergent integral in the limit $\epsilon \to 0$, because

$$\mathrm{Re} \int_0^1 \mathrm{d}\omega \left| \frac{1}{a - \omega + \mathrm{i}\epsilon} \right|^2 = \frac{\pi}{\epsilon} + \frac{1}{a(a-1)} + \frac{1}{3} \left(\frac{1}{a^3} - \frac{1}{(a-1)^3} \right) \epsilon^2 + \mathcal{O}(\epsilon^4) \,. \tag{10}$$

An infinite two-photon decay rate cannot be considered physically sensible.

We can thus conclude that virtual intermediate states with energies that lie between the energy of the initial and final states of a two-photon decay process contribute a finite correction to the two-photon decay rate, although the integrand of Eq. (8), in the limit $\epsilon \to 0$, has a non-integrable singularity at $\omega = a$ of the form $1/(a-\omega)^2$. The convergence of the integral (8) in the limit $\epsilon \to 0$ is not uniform, and our example shows that the concept of non-uniform convergence is not merely a mathematical sophistication in the context of two-photon processes: it ensures that finite, physically sensible results are obtained for the two-photon decay rates, which include the contribution from all possible virtual states. A generalization of the results obtained in this communication to the relativistic and to the many-electron case is straightforward. Finally, it should be remarked that at least in principle, the two-photon decay rates can be measured experimentally although they are orders of magnitude smaller than one-photon rates for many of the processes listed in Table 1. The point is that only in two-photon decay, both photons can be detected in coincidence, and events can be selected by considering the center-of-mass energy.

The author acknowledges support from Deutsche Forschungsgemeinschaft (Heisenberg program).

References

[1] A. J. Duncan, Z. A. Sheikh, H.-J. Beyer, and H. Kleinpoppen, J. Phys. B **30**, 1347 (1997).

[2] R. Barbieri and J. Sucher, Nucl. Phys. B **134**, 155 (1978).

[3] U. D. Jentschura, Phys. Rev. A **69**, 052118 (2004).

[4] V. Florescu, Phys. Rev. A **30**, 2441 (1984).

[5] J. D. Cresser, A. Z. Tang, G. J. Salamo, and F. T. Chan, Phys. Rev. A **33**, 1677 (1986).

[6] V. Florescu, I. Schneider, and I. N. Mihailescu, Phys. Rev. A **38**, 2189 (1988).

[7] K. Pachucki, Phys. Rev. A **63**, 042503 (2001).

[8] U. D. Jentschura, J. Evers, C. H. Keitel, and K. Pachucki, New J. Phys. **4**, 49 (2002).

[9] U. D. Jentschura, Phys. Rev. A **74**, 062517 (2006).